

Appln. No. 10/706,645
Amendment dated January 29, 2007
Reply to Final Office Action of November 28, 2006

Amendments to the Claims

This listing of claims will replace all prior versions, and listings, of claims in the application:

Listing of Claims:

1. (Previously presented) A high temperature stable catalyst support comprising the following crystalline structures:
 - an alumina phase selected from the group consisting of alpha-alumina, theta-alumina and combinations thereof;
 - a rare earth aluminate of a hexaaluminate or hexaaluminate-like structure comprising at least one rare earth metal and having a high molar ratio of aluminum to rare earth metal between 11:1 and 14:1; and
 - a rare earth aluminate of a perovskite or perovskite-like structure comprising the at least one rare earth metal and having a low molar ratio of aluminum to rare-earth metal of less than 2:1,wherein the catalyst support contains not more than 20% of alpha-alumina, and less than 100 wt% to about 1 wt% of the rare earth aluminate with the high molar ratio of aluminum to rare earth metal.
2. (Previously presented) The catalyst support of claim 1 wherein the catalyst support comprises between 5 and 50 percent by weight of the rare earth aluminate with the high molar ratio of aluminum to rare earth metal based on the total weight of the catalyst support.
3. (Previously presented) The catalyst support of claim 1 wherein the at least one rare earth metal is selected from the group consisting of lanthanum, neodymium, praseodymium, cerium, samarium, and combinations thereof.
4. (Previously presented) The catalyst support of claim 1 wherein both rare earth aluminates comprise lanthanum.
5. (Previously presented) The catalyst support of claim 1 wherein the catalyst support comprises between about 1 wt% and about 10 wt% of lanthanum as the at least one rare earth metal.

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6. (Previously presented) The catalyst support of claim 1 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal further comprises an element from Groups 1-14 of the Periodic Table of Elements.
7. (Previously presented) The catalyst support of claim 1 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal further comprises nickel, magnesium, barium, potassium, sodium, manganese, a second rare earth metal or combinations thereof.
8. (Previously presented) The catalyst support of claim 1 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal and the alumina phase are intimately mixed.
9. (Previously presented) The catalyst support of claim 1 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal coats the alumina phase partially or completely.
10. (Canceled)
11. (Previously presented) The catalyst support of claim 1 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal has a chemical formula of LnAl_yO_z , where y is between 11 and 14; and z is between 18 and 23, Al and O represent aluminum atoms and oxygen atoms respectively; and where Ln comprises lanthanum, neodymium, praseodymium, samarium, or combinations thereof.
12. (Previously presented) The catalyst support of claim 1 wherein the rare earth aluminate with the high molar ratio of aluminum to rare-earth metal has a chemical formula of MAl_yO_z , where y is between 11 and 12; z is between 18 and 19; Al and O represent aluminum atoms and oxygen atoms respectively; and M comprises a combination of lanthanum and samarium.
13. (Previously presented) The catalyst support of claim 1 wherein the rare earth aluminate with the high molar ratio of aluminum to rare-earth metal comprises a lanthanum hexaaluminate.
- 14-15. (Canceled)
16. (Previously presented) The catalyst support of claim 1 further comprising an oxide of the at least one rare earth metal.

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17. (Original) The catalyst support of claim 1 wherein the catalyst support has a surface area greater than $2 \text{ m}^2/\text{gram}$.
18. (Original) The catalyst support of claim 1 wherein the catalyst support has a surface area lower than $30 \text{ m}^2/\text{gram}$.
19. (Previously presented) The catalyst support of claim 1 wherein the catalyst support comprises between 1 wt% and 10 wt% of the at least one rare earth metal.
20. (Previously presented) The catalyst support of claim 1 wherein the catalyst support is made by impregnating a solution of [[a]] the at least one rare earth metal onto an aluminum-containing precursor; and calcining at a temperature between $1,000^\circ \text{C}$ and $1,600^\circ \text{C}$.
21. (Original) The catalyst support of claim 20 wherein the aluminum-containing precursor comprises an aluminum structure selected from the group consisting of bayerite, gibbsite, boehmite, pseudo-boehmite, bauxite, gamma-alumina, delta-alumina, chi-alumina, rho-alumina, kappa-alumina, eta-alumina, theta-alumina, and combinations thereof.
22. (Original) The catalyst support of claim 20 wherein the aluminum-containing precursor comprises at least one transition alumina selected from the group consisting of gamma-alumina, delta-alumina, chi-alumina, rho-alumina, kappa-alumina, eta-alumina, and theta-alumina.
23. (Previously presented) The catalyst support of claim 20 wherein the aluminum-containing precursor comprises gamma-alumina.
24. (Previously presented) The catalyst support of claim 23 wherein calcining is done at a temperature between about $1,200^\circ \text{C}$ and about $1,300^\circ \text{C}$.
25. (Original) The catalyst support of claim 23 wherein calcining is done at a temperature between $1,100^\circ \text{C}$ and $1,400^\circ \text{C}$.
26. (Withdrawn) A method for making a thermally stable aluminum-based catalyst support comprising:
 - (a) impregnating a solution of a rare earth metal onto an aluminum-containing

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precursor;

- (b) drying the impregnated aluminum-containing precursor; and
- (c) calcining at a temperature between 1,000 °C and 1,600 °C in a manner effective to convert a portion of said aluminum-containing precursor to an aluminum oxide phase comprising alpha-alumina, theta-alumina, or combinations thereof, and to convert another portion of said aluminum-containing precursor with at least a fraction of said rare earth metal to a rare earth aluminate of a hexaaluminate or hexaaluminate-like structure with a high molar ratio of aluminum to rare earth metal between 11:1 and 14:1, and further to a second rare earth aluminate of a perovskite or perovskite-like structure with a low molar ratio of aluminum to rare earth metal of less than 2:1,

such that the catalyst support comprises said rare earth aluminate with the low molar ratio of aluminum to rare-earth metal; the aluminum oxide phase comprising less than 20 wt% of alpha-alumina; and less than 100 wt% to about 1 wt% of said rare earth aluminate with the high molar ratio of aluminum to rare earth metal.

27. (Withdrawn) The method of claim 26 wherein the aluminum-containing precursor comprises an aluminum structure selected from the group consisting of bayerite, gibbsite, boehmite, pseudo-boehmite, bauxite, gamma-alumina, delta-alumina, chi-alumina, rho-alumina, kappa-alumina, eta-alumina, theta-alumina, and combinations thereof.
28. (Withdrawn) The method of claim 26 wherein the aluminum-containing precursor comprises a transition alumina selected from the group consisting of gamma-alumina, delta-alumina, chi-alumina, rho-alumina, kappa-alumina, eta-alumina, theta-alumina, and combinations thereof.
29. (Withdrawn) The method of claim 26 wherein the aluminum-containing precursor comprises gamma-alumina.
30. (Withdrawn) The method of claim 29 wherein calcining is done at a temperature between about 1,200 °C and about 1,300 °C.
31. (Withdrawn) The method of claim 29 wherein calcining is done at a temperature between

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1,100 °C and 1,400 °C.

32. (Withdrawn) The method of claim 26 wherein the rare earth metal is selected from the group consisting of lanthanum, neodymium, praseodymium, cerium and combinations thereof.
33. (Withdrawn) The method of claim 26 wherein both rare earth aluminates comprise lanthanum.
34. (Withdrawn) The method of claim 26 wherein calcining is further effective to further convert another portion of the rare earth metal solution into an oxide of said rare earth metal, said rare earth oxide consisting essentially of rare earth metal atoms and oxygen atoms.
35. (Withdrawn) The method of claim 26 wherein the solution of rare earth metal comprises more than one rare-earth metal.
36. (Canceled)
37. (Withdrawn) The method of claim 26 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal comprises a lanthanum hexaaluminate.
38. (Withdrawn) The method of claim 26 wherein the catalyst support comprises between about 0.5 and about 10 percent by weight of the rare earth metal based on the total weight of the catalyst support.
39. (Withdrawn) The method of claim 26 wherein the catalyst support comprises between about 5 and about 50 percent by weight of the rare earth aluminate with the high molar ratio of aluminum to rare earth metal based on the total weight of the catalyst support.
40. (Currently amended) A partial oxidation catalyst comprising:
- (a) an active ingredient comprising a rhodium alloy or a metal selected from the group consisting of rhodium, iridium, ruthenium and combinations thereof, wherein, when the active ingredient comprises rhodium, said rhodium being is in an amount of from about 0.1 wt% to about 20 wt% based on the total catalyst weight; and
 - (b) a support onto which the active ingredient is dispersed, said support comprising

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an alumina phase selected from the group consisting of alpha-alumina, theta-alumina and combinations thereof;

a rare earth aluminate of a hexaaluminate or hexaaluminate-like structure comprising a rare-earth metal and having a high molar ratio of aluminum to rare earth metal between 11:1 and 14:1; and

a rare earth aluminate of a perovskite or perovskite-like structure comprising the rare-earth metal and having a low molar ratio of aluminum to rare earth metal of less than 2:1;

wherein the support comprises less than 100 wt% to about 1 wt% of said rare earth aluminate with the high molar ratio of aluminum to rare earth metal based on the total weight of the support.

41. (Canceled)

42. (Previously presented) The partial oxidation catalyst of claim 40 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal comprises a lanthanide hexaaluminate, and the lanthanide metal is selected from the group consisting of lanthanum, neodymium, praseodymium, samarium, and combinations thereof.

43. (Original) The partial oxidation catalyst of claim 42 wherein the lanthanide hexaaluminate comprises a lanthanide metal selected from the group consisting of lanthanum, neodymium, praseodymium, and combinations thereof.

44. (Original) The partial oxidation catalyst of claim 42 wherein the lanthanide hexaaluminate comprises lanthanum.

45. (Original) The partial oxidation catalyst of claim 40 wherein the rare earth metal is selected from the group consisting of lanthanum, neodymium, praseodymium, and combinations thereof.

46. (Previously presented) The partial oxidation catalyst of claim 40 wherein both rare earth aluminates comprise lanthanum.

47. (Previously presented) The partial oxidation catalyst of claim 40 wherein the rare earth

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aluminate with the high molar ratio of aluminum to rare earth metal comprises more than one rare-earth metal.

48. (Original) The partial oxidation catalyst of claim 40 wherein the support has a surface area greater than $2 \text{ m}^2/\text{gram}$.

49. (Previously presented) The partial oxidation catalyst of claim 40 wherein the support comprises between about 5 and about 50 percent by weight of the rare earth aluminate with the high molar ratio of aluminum to rare-earth metal based on the total weight of the support.

50. (Withdrawn) A method for making synthesis gas comprising:
converting a gaseous hydrocarbon stream and an oxygen-containing stream over a partial oxidation catalyst, to make a product stream comprising CO and H₂,

wherein said partial oxidation catalyst includes

an active ingredient comprising a rhodium alloy or a metal selected from the group consisting of rhodium, iridium, ruthenium, and combinations thereof, wherein, when the active ingredient comprises rhodium, said rhodium being-is in an amount of from about 0.1 wt% to about 20 wt% based on the total catalyst weight; and

a support onto which the active ingredient is dispersed, said support comprising

an alumina phase selected from the group consisting of alpha-alumina, theta-alumina and combinations thereof;

a rare earth aluminate of a hexaaluminate or hexaaluminate-like structure comprising a rare earth metal and having a high molar ratio of aluminum to rare earth metal between 11:1 and 14:1; and

a rare earth aluminate of a perovskite or perovskite-like structure comprising the rare-earth metal and having a low molar ratio of aluminum to rare earth metal of less than 2:1;

wherein the support comprises less than 100 percent to about 1 percent by weight of said rare earth aluminate with the high molar ratio of aluminum to rare earth metal based on the total weight of the support.

51. (Withdrawn) The method of claim 50 wherein the support comprises between about 1 and

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about 50 percent by weight of said rare earth aluminate with the high molar ratio of aluminum to rare earth metal based on the total weight of the support.

52. (Withdrawn) The method of claim 50 wherein the rare earth metal is selected from the group consisting of lanthanum, neodymium, praseodymium, cerium, and combinations thereof.
53. (Withdrawn) The method of claim 50 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal comprises lanthanum.
54. (Withdrawn) The method of claim 50 wherein the support comprises between about 1 and about 10 percent by weight of lanthanum based on the total weight of catalyst support.
55. (Withdrawn) The method of claim 50 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal comprises lanthanum and samarium.
56. (Canceled)
57. (Withdrawn) The method of claim 50 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal and the alumina phase are intimately mixed.
58. (Withdrawn) The method of claim 50 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal coats the alumina phase partially or completely.
59. (Canceled)
60. (Withdrawn) The method of claim 50 wherein the rare earth aluminate with the high molar ratio of aluminum to rare earth metal comprises a lanthanum hexaaluminate.
61. (Withdrawn) The method of claim 50 wherein the support comprises between about 1 wt% and about 10 wt% of the rare earth metal.
62. (Withdrawn) The method of claim 50 wherein the conversion is done at a GHSV between about 20,000 hr⁻¹ to about 100,000,000 hr⁻¹.
63. (Withdrawn) The method of claim 50 wherein the conversion is done at a temperature

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between about 350 °C to about 2,000 °C.

64. (Withdrawn) The method of claim 50 wherein the conversion is done at a pressure between about 100 kPa to about 4,000 kPa.
65. (Withdrawn) The method of claim 50 wherein the hydrocarbon stream comprises natural gas.
66. (Withdrawn) The method of claim 50 wherein the hydrocarbon stream comprises at least about 50% by volume of methane.
67. (Withdrawn) The method of claim 50 wherein the partial oxidation catalyst comprises rhodium.
68. (Withdrawn) The method of claim 67 wherein the partial oxidation catalyst comprises between about 0.5 wt% and about 10 wt% of rhodium.
69. (Withdrawn) The method of claim 67 wherein the partial oxidation catalyst has a rhodium surface area greater than about 0.5 m²/g.
70. (Withdrawn) The method of claim 67 wherein the partial oxidation catalyst further comprises samarium.
71. (Withdrawn) The method of claim 50 wherein the catalyst exhibits hydrocarbon conversion of equal to or greater than about 85%.
72. (Withdrawn) The method of claim 50 wherein the partial oxidation catalyst has a hydrogen selectivity equal to or greater than about 85%.
73. (Withdrawn) The method of claim 50 wherein the product stream comprising CO and H₂ has a H₂:CO molar ratio between about 1:4:1 and 2.3.
74. (Withdrawn) The method of claim 50 wherein at least a portion of the product stream comprising CO and H₂ is further converted to hydrocarbons.

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75. (Canceled)
76. (Previously presented) The catalyst support of claim 1 wherein the catalyst support comprises between 1 and 50 percent by weight of the rare earth aluminate with the high molar ratio of aluminum to rare earth metal based on the total weight of the catalyst support.
77. (Previously presented) The catalyst support of claim 1 wherein the catalyst support comprises more than 40 wt% and less than 100 wt% of the rare earth aluminate with the high molar ratio of aluminum to rare earth metal based on the total weight of the catalyst support.
78. (Withdrawn) The method of claim 26 wherein the catalyst support comprises between about 1 and about 50 percent by weight of the rare earth aluminate with the high molar ratio of aluminum to rare earth metal based on the total weight of the catalyst support.
79. (Withdrawn) The method of claim 26 wherein the catalyst support comprises more than 40 wt% and less than 100 wt% of the rare earth aluminate with the high molar ratio of aluminum to rare earth metal
80. (Previously presented) The partial oxidation catalyst of claim 40 wherein the support comprises between about 1 and about 50 percent by weight of the rare earth aluminate with the high molar ratio of aluminum to rare earth metal based on the total weight of the support.
81. (Previously presented) The partial oxidation catalyst of claim 40 wherein the support comprises more than 40 wt% and less than 100 wt% of the rare earth aluminate with the high molar ratio of aluminum to rare earth metal based on the total weight of the support.
82. (Canceled)
83. (Previously presented) The catalyst support of claim 40 wherein the support comprises not more than about 20 wt% alpha-alumina.
84. (Previously presented) The catalyst of claim 40 wherein the partial oxidation catalyst comprises a rhodium alloy, said rhodium alloy comprising a metal selected from the group

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consisting of ruthenium, iridium, platinum, palladium, tantalum, niobium, molybdenum, rhenium, tungsten, cobalt, and zirconium.

85. (Previously presented) The catalyst of claim 40 wherein the partial oxidation catalyst comprises between about 0.5 wt% and about 10 wt% of rhodium.

86. (Previously presented) A partial oxidation catalyst comprising:

(a) an active ingredient comprising rhodium or a rhodium alloy, said rhodium alloy comprising rhodium and a metal selected from the group consisting of ruthenium, iridium, platinum, palladium, tantalum, niobium, molybdenum, rhenium, tungsten, cobalt, and zirconium, said rhodium being in an amount of from 0.1 wt% to about 20 wt% based on the total catalyst weight; and

(b) a support onto which the active ingredient is dispersed, said support comprising:
an alpha-alumina phase;

less than 100 percent to about 1 percent by weight of a rare earth aluminate of a hexaaluminate or hexaaluminate-like structure, with a high molar ratio of aluminum to rare-earth metal between 11:1 and 14:1, and comprising at least one rare earth metal selected from the group consisting of lanthanum, neodymium, praseodymium, samarium, and combinations thereof; and

a rare earth aluminate of a perovskite structure comprising the at least one rare earth metal with a low molar ratio of aluminum to rare-earth metal of less than 2:1.

87. (Previously presented) The partial oxidation catalyst of claim 86 wherein the rare earth aluminate with the high molar ratio of aluminum to rare-earth metal comprises between 5 and 45 percent by weight of the total weight of the catalyst support.

88. (Previously presented) The partial oxidation catalyst of claim 86 wherein the rare earth aluminate with the high molar ratio of aluminum to rare-earth metal comprises more than 40 percent and less than 100 percent by weight of the total weight of the catalyst support.

89. (Previously presented) The partial oxidation catalyst of claim 86 wherein the partial oxidation catalyst comprises between about 0.5 wt% and about 10 wt% of rhodium.

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90. (Previously presented) The partial oxidation catalyst of claim 86 wherein the partial oxidation catalyst comprises rhodium and has a rhodium surface area greater than about 0.5 m²/g rhodium.